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Helium pressure increase inside GPHS clads: implications for formation of distended clads

Roberta N. Mulford

Distension of GPHS clads has generally been attributed to gas buildup inside the clad. The bulging vent frit cover can be caused only by gas pressure, since the vent cover is isolated from the fuel by the vent frit, as shown in Figure 1. Gas generation is the only change expected as the oxide fuel ages. However, several data suggest that gas pressure is not the origin of the distension of the clad body. Gas pressure sufficient to distend the iridium body of the clad would be on the order of 1500 psi. [1] This high pressure is very unlikely to be reached inside a GPHS clad, because the vent frit cover weld cannot sustain high pressures. [2] Every distended clad that has been examined has an open vent frit, suggesting that the vent frit cover eventually opens on all distended GPHS clads. No gas pressure has been observed in any distended clad. [3] In addition, the vent frit cover is sufficiently thin, about 2 grains thick, that diffusion of helium through the intact cover is possible and almost certainly occurs. [4]

Conversely, no clad has ever been definitively observed to distend after venting. Removal of the vent cover allows free escape of any radiolytically generated helium. However, there is little data on the behavior of vented GPHS. The vent cover is seldom removed at LANL, and vent covers are not removed on clads that will be stored. The vent covers are known to have been removed from the four clads residing in a module which was stored for 8 years, shown in Figure 3.

Expansion of the fuel is more likely to provide sufficient force on the iridium clad to distort the iridium metal. Radiographs indicate that the fuel inside a distended GPHS clad is also distended, filling all or most of the interior volume of the GPHS, as shown in Figure 2. No radiograph shows the fuel expanded and in contact with the weld shield and the distorted side of the GPHS clad without distension. [3] However, radiography is infrequently done on stored clads unless they exhibit distension, so the dimensions of fuel in normal clads are not generally known.

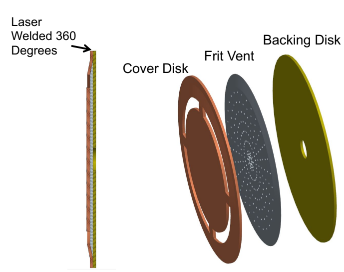
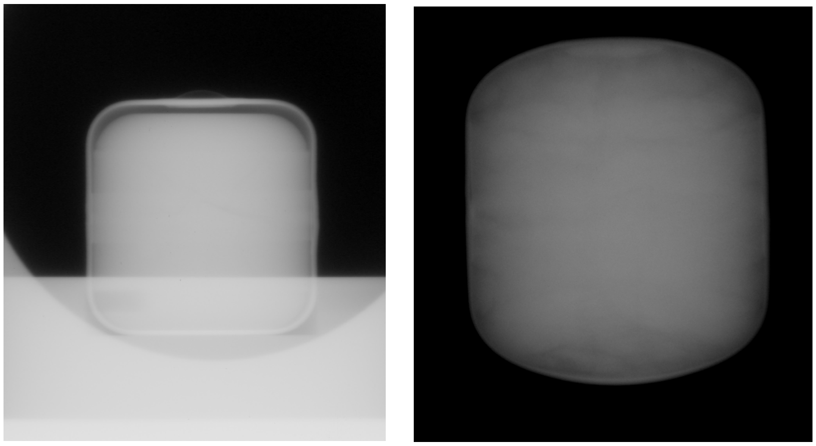


Figure 1. exterior view of distended clad vent cover, and detail of vent frit.
The vent cover weld encircles the vent and vent cover assembly.



2a.) 2b.)

Figure 2. Typical radiographs, showing a.) normal pellet and b.) distended fuel

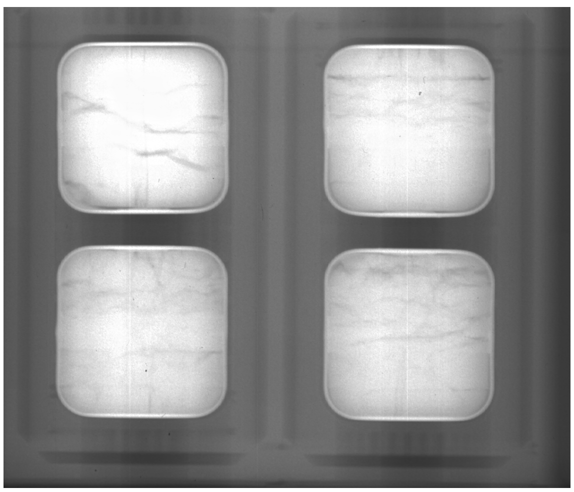


Figure 3. Radiographs of vented clads stored in the SVT-14 module. Cracks can be seen in the fuel.

In an effort to prevent the development of any new distended clads, removal of the vent frit cover in now recommended, to allow free venting of any helium produced by the fuel in the clad. Venting is recommended once the clad has reached 18 months of age to prevent clad pressurization by helium. Gas sampling of distended clads has demonstrated that all clads examined to date are at ambient pressure, and contain air. While venting the clad probably does little harm, it may serve only to supplement the spontaneous vent frit cover rupture that has been observed to occur.

While no mechanism for clad distension has been definitively demonstrated, several phenomena may be contributing to expansion of the fuel.

When encapsulation is complete, the vent cover is an integral sealed enclosure, and the fuel resides in a closed volume. As the PuO_2 undergoes decay, helium is generated in this closed volume. Helium accumulates in the closed volume of the clad, in pore space in the fuel pellet, and probably also in bubbles that form within the fuel. [5] At 81% TMD, the fuel has unoccupied volume inside it. This pore space is deliberately incorporated into the fuel in order to accommodate helium. No SEM images of the fuel in distended clads exist, so postulating the presence of bubbles in the fuel relies on historical data. SEM images of fuel obtained between 1974 and 1984 show bubbles within grains and at grain boundaries in the fuel. [5, 6] Helium also builds up outside the fuel, in the ullage, the free space between the fuel and the iridium cladding. The partitioning of the helium between the pore space and the free volume inside the fuel is not known, but pressure equilibrium can reasonably be anticipated between the free volume inside the fuel and the ullage between the fuel and the encapsulation.

There is evidence that helium builds up in the fuel, within the matrix. [6, 7, 8, 9] As much as 75% of the helium inventory has been observed to remain in the fuel until it is heated. [6, 7, 8, 9] In principle, all of the radiolytic helium can be incorporated into the oxide matrix, but in practice, much of the helium is apparently released from the fuel. Partitioning of the evolved helium between the fuel and the available space inside the clad is not known.

This report includes several rough calculations to examine the action of helium gas on the fuel, and the role that may be played by pressure in the formation of a distended clad. The quantity of helium available as a function of the age of the clad is easily calculated from the isotopic composition of the fuel and the time elapsed since encapsulation of the pellet. The ullage volume between the clad wall and the fuel is 1667 mm^3 as determined from CAD drawings, and 668.4 mm^3 as measured from radiographs of a new and undistended clad, FCO551. [10] The pellet volume is known, and the pore volume within the pellet can be calculated from the pellet density. Volumes are listed in Table 1.

Helium builds up within the fuel. There is no reason for helium to leave the fuel as long as the pressure inside the GPHS clad and the pressure outside the pellet are the same, and the vent cover is intact. As helium is generated, it fills the free space until the volume within the pellet and the clad is occupied. Two cases are presented below, helium occupying only pore volume and helium occupying the sum of ullage and pore volume.

Helium inside the fuel apparently continues to increase until some threshold is reached: some “on-switch” for distension of the fuel. Two separate cases should be considered in thinking about distended clads. Clads that have been exposed to high temperatures differ from clads that have distended during normal storage. Radiographic images of the fuel in clads that have gotten hot show distinct differences from images of fuel in old clads that are distended as seen in Figure 4. Although clear records do not exist to confirm that very old clads have not experienced thermal excursions, it is likely that they have distended without undergoing heating. (Figure 4b)

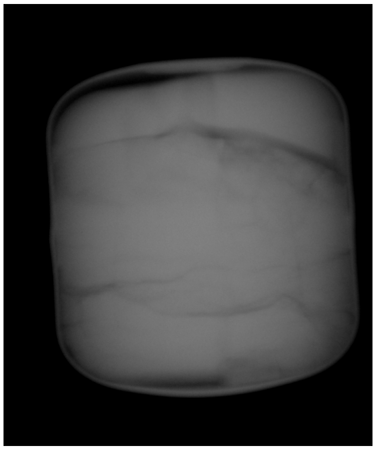
Thermal excursion clads

Clads that have reached high temperatures during storage may be expected to experience sudden pressurization due to release of helium dissolved within the fuel. [6] When fuel in a clad is reaches a temperature above 1425°C, all helium entrained in the fuel is released into the gas phase, [7] Gas release accelerates as the fuel temperature increases from ambient, and becomes noticeable above about 900°C. [11]

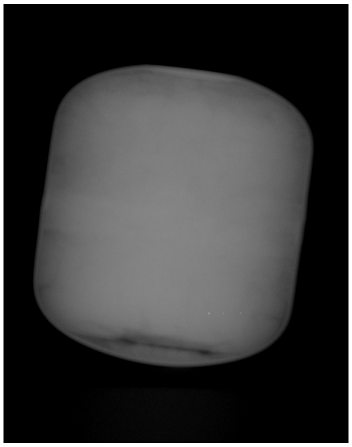
Fuel after a thermal excursion, seen in the radiographic image in Figure 4a, exhibits large cracks and open volume within the fuel. Clads distended after thermal excursions have a few large cracks and the fuel is in a few large fragments. Measured fragment sizes are shown in Figures 5 and 6. In addition, examination of examples of this fuel shows a distinct lack of fragments smaller than 5 mm relative to other distended clads examined as shown in Figures 5 and 6a. [12] In the case of a clad that has been subject to a thermal excursion, it is reasonable to expect the thermal perturbation to be the precipitating factor for distension.

Fractured fuel is a common feature seen in distended clads that have experienced elevated temperatures. The fracturing is most likely a result of thermal stresses incurred during a thermal excursion, although it may be a feature of a pellet that was fractured at the time of encapsulation. Fractured fuel is observed in the four vented clads stored at a relatively higher temperature in the SVT-14 module, as seen in Figure 3.

Historically, temperature excursions were identified as the cause of all cases of distension. Accordingly, clads in containers are stored in a cold water bath and are constrained to one clad per container. If not in the water bath, clads are stored with adequate spacing to preclude excessive mutual heating.



4a.)



4b.)

Figure 4: Examples of radiographs of distended clads: 4a.) FCO 371, distended after thermal excursion, and 4b.) FCO 421, distended without thermal excursion.

Particle size fractions have been measured for the fuel from eight of the ten identified distended clads. Fragment sizes shown in Figure 5 fall into three major groups. Clads known to have experienced thermal excursions exhibit large fragments, shown in Figure 6a. Smaller fragments and two distinct behaviors are evident in clads that distended during storage at ambient temperature, shown in Figure 6b.

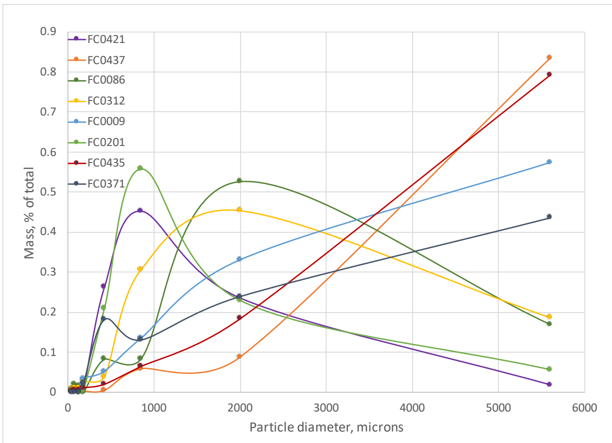
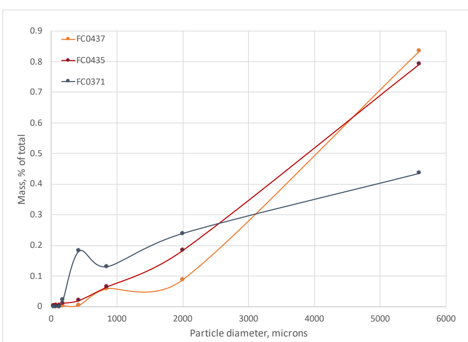
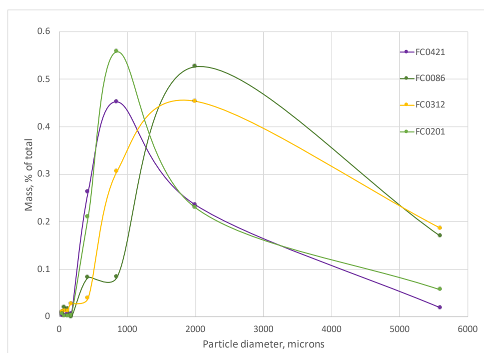


Figure 5. Particle sizes measured in fuel from several distended clads. [12]





6a.) 6b.)

Figure 6. Particle sizes for a) clads known to have experienced thermal excursions and exhibiting primarily large fragments. b) clads that distended during storage at ambient temperature.

<u>Ambient temperature clads</u>

For a GPHS that has most likely distended without a thermal excursion, some critical age or condition must exist that results in distension. There are several possibilities. Bubbles in the fuel may reach a pressure that exceeds the yield strength of the fuel, resulting in prompt local fuel fracture. Helium pressure within bubbles or pore space within the fuel may reach some critical value related to a factor other than yield strength, precipitating fuel expansion. If the vent cover weld breaches, the resulting low pressure outside the fuel may provide a thermodynamic driver for expansion of helium within fuel, possibly expanding the volume of the pellet, where previously the pressure equilibrium between the interior and exterior of the pellet allowed gas to be stable at high pressures in the fuel. Reducing the pressure around the fuel may provide a thermodynamic driver for exit of helium from fuel. Some other phenomenon may be precipitating fuel swelling and distension of the iridium encapsulation of the GPHS.

Fractures in the fuel pellet are visible in both thermally driven distension and distension at ambient temperature. Pellets distending at ambient temperature have a myriad of small closed cracks. (Figure 4b, FC0421) The fuel fragment size appears to reflect the dimension and number of cracks. Clads that became distended at ambient temperature have many hairline cracks and accordingly exhibit fine particles, as shown in Figures 4, 5, and 6.

Fragment sizes appear to be similar for clads sharing common distension behavior, within the few examples examined (Figure 5.) It is possible that the two patterns of distension share a common mechanism, and differ in the size and number of cracks that develop.

<u>The vent cover</u>

The vent cover (Figure 1) is put on the iridium clad before fabrication of the GPHS, and serves to protect the vent from infiltration and damage by the acid bath used to wash the exterior of the clad free of contamination after welding. The iridium vent cover is about two grains thick, probably making the grain boundaries somewhat permeable to helium. The weld between the vent cover and the iridium clad body is the weak point in the vent cover, and vent cover weld rupture has been observed during dynamic pressurization of the vent assembly. [2]

The vent cover gives way at the weld. [2] The pressure at which vent cover weld breach occurs under gradual pressurization, i.e. quasi-static loading, has not been measured. Under dynamic loading by abrupt pressurization, the measured values at which the vent cover weld ruptured are between 150 and 180 psig. [2]

An open vent cover allows the free space around the fuel to vent to the atmosphere, and creates a pressure differential between the inside of fuel and the ullage, or between bubbles and the ullage.

Calculations examine how several factors may influence the pressure anticipated within the clad.
1 helium accumulation in the porosity and the ullage
2 Yield strength of the fuel
3 Yield strength of the vent cover

Several important parameters affecting fuel expansion are not known. The yield strength of the fuel in tension is not known. Reported values were measured in compressive loading and are likely too high. [13] The maximum gas pressure that can be achieved before the vent cover opens under gradual pressurization is not known, although the strength of the vent cover weld under sudden pressurization has been measured. [2] The ullage is observed to decrease as the pellet ages. [3, 10] The partitioning of helium between fuel, porosity, and ullage is not known.

Table 1 Clad dimensions, Distended and Normal

	Volume, liters	source	Refc.
interior of clad	0.0182		3, 10
volume of fuel	0.0163 liters	in 421 originally	3
	0.0165	in 201	3
	0.0159 to 0.0169 liters.	From drawings	
Calculated pore volume inside fuel at 81-82% TMD	0.003135 liters.	0.0165 x 0.19	
volume of the ullage	0.001667 liters	calculated from CAD drawings	10
	0.0006684 liters	measured on FCO551 radiographs	10
total volume, ullage + pore space	0.004802 liters	from CAD drawings	
	0.0038034 liters	measured on FCO551	

Calculations are performed at two temperatures. The first is the temperature 841°C, calculated at the center of a pellet stored in a configuration known to cause distension. [1] The internal fuel temperature of 600°C is also used in calculations.

Table 2 Temperatures

T, °C	source	Refc
841	pellet internal temperature	11
600	canonical pellet temperature	

Helium inventory over the pellet age

The spreadsheet PUDECAY was used to determine helium inventories. This spreadsheet calculates helium output from all isotopes in a sample and from daughter products. The helium output from 152 grams of $^{238}PuO_2$ fuel at various times is tabulated.

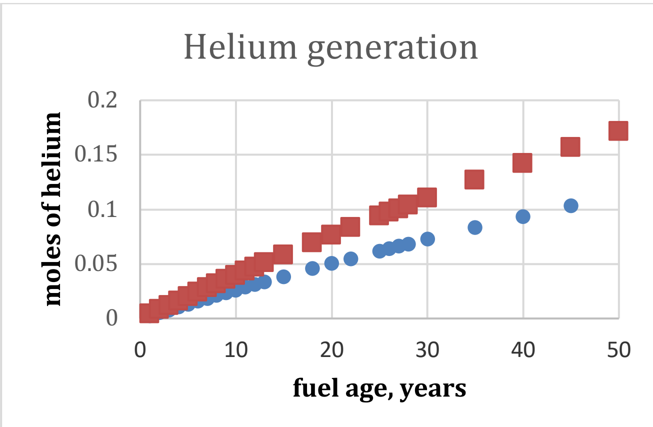


Figure 7. Helium inventory in the clads as a function of time.

Calculation 1, helium filling available porosity and ullage

Helium will accumulate until it fills the available porosity. If helium can leave the pellet, it accumulates until it fills both the porosity and the ullage between the pellet and the clad. Consider a case in which the PuO_2 pellet has no strength. As soon as helium is generated, the fuel would begin to expand. The timescale for increase in the pellet volume is calculated.

To fill the ullage calculated from the CAD drawing, accumulation of 0.001667 liters of gas within the pellet is required.
At 841°C, PV/RT = 1*0.001667/(0.08260*1114K) = 1.82 E-5 moles of gas inside pellet, which is generated in 2 or 3 days. If the temperature is 600°C, the quantity is 2.33E-5 moles, generated in about 3 days. If the FC0551 ullage volume of 0.0006684 liters is used, expansion of the pellet to fill the available space takes about a day. The free volume in the GPHS is assumed to be at about an atmosphere initially, but if the fuel starts out in a vacuum, then filling the free volume in the fuel and the ullage inside the cladding would require only a few days, no more than 5.

If the initially generated helium became entrained within the fuel, 75% of the helium would be consumed. It would then require 20 days at 600°C to fill up the pellet, the porosity, and the ullage, or 15 days at 841°C.

Radiographs indicate that pellet expansion does not occur on this short timescale. The pellet has finite strength, which prevents immediate expansion. Without strength or confinement to prevent helium from expanding the fuel, the pellet could double in size in a month.   Distension of either pellet or clad is not observed on this short timescale, and the strength of the PuO_2 pellet is a necessary term in the calculation.

<u>Observed helium inventories</u>

Pressure has been measured to be small inside of all of the distended clads. [3, 12] Only a small fraction of the helium that has been generated is present in the measured clads. In addition, air is frequently observed in the clads. [3, 12] For example, FC0421 was gas sampled and determined to contain only 9.72E-4 moles of helium, occupying both pore space and ullage. This amount of helium is generated in about 2.8 months, and is a small fraction of the inventory anticipated after the 5.7 years of helium generation that preceded the gas sampling of FCO421. [3]

<u>Strength of PuO_2.</u>

The strength of the fuel prevents the fuel from distending until some threshold pressure is reached. Radiographs (e.g. FC0511, shown in Figure 8) suggest that the pellet retains its size and shape for some time after encapsulation. Disassembled clads generally corroborate this information, with the pellet sliding out of the clad.

Figure 8. Radiograph of FC0511 after 55 days of aging.

The available measurements of fuel strength were made in compression [13] whereas bubbles or pressurized pores exert tensile stress. The tensile strength of ceramics is generally extremely low, but in the absence of other data, the compressive data gives a good first estimate. [13] At temperatures of 1100-1150°C, the PuO_2 fuel exhibits yield at about 150 MPa. At 900°C, a stress of 200 MPa is reached before the fuel fractures. No data at temperatures below 900°C is reported. [13]

The PuO_2 pellet is a “simple oxide” ceramic, most similar to alumina and other similar oxides used in refractory applications. The tensile strength of these ceramics is almost entirely determined by flaws. Fracture is usually trans-granular, relying on cleavage planes rather than on intergranular fracture. Micro-cracks effect toughening of the ceramic, by redirecting energy at the crack tip, or "blunting." This suggests that the initial fracture arising from helium ingrowth might serve to strengthen the material, and that the tensile yield value might actually increase with time. [14]

<u>Calculation 2</u>, PuO_2 has strength, assume compressive yield governs distension

Suppose the fuel can hold pressure in bubbles to the compressive yield strength of 150 MPa or to 200 MPa, and then yields and cracks. 200 MPa is reached at a pressure of 1974 atm, and 150 MPa at 1455 atm.

The number of moles required to reach 1974 atm in the 0.003135-liter pore volume is calculated at both 600°C and 841°C. This volume is the volume of the porosity inside the pellet, and does not include the ullage outside the pellet.

At 841°C, PV/RT = (1974*0.003135)/(0.08206*1114) = 6.77 * 10^{-2} moles. At 841°C, this is 17.5 years' accumulation in 152 grams of fuel. At a temperature of 600°C, 8.64E-2 moles are necessary, which requires 22 years to fill the pore volume. If helium is allowed to occupy both the pores and the ullage, the time to reach 1974 atm of pressure increases to 45 years at either temperature.

To reach the lower yield strength of 150 MPa,
PV/RT = (1455*0.003135)/(0.08206*1114) = 5 * 10^{-2} moles, or about 12.5 years' accumulation at 841°C, or 16.5 years accumulation at 600°C (6.37E-2 moles). If the ullage is included, 26 years are required at 600°C, or 20 years at 841°C. If helium is entrained in the lattice, then the time required to reach the required pressure is much longer.

The youngest distended clads observed to date, FCO462 and FCO463, appear to have become distended within 3 years of encapsulation. These two clads distended after storage at elevated temperatures. Distension without heating (e.g. FCO421) distension has been observed within 6 years. These timescales are shorter than the 12 to 45 years calculated under the hypothesis that the [compressive] strength of the fuel is the determining factor in the onset of fuel distension.

If helium is entrained in the fuel and the clad is heated above the helium release temperature, the helium can be released abruptly from the fuel by heating. [6, 7] At the helium release temperature of 1400°C, the time required to accumulate sufficient helium pressure is reduced to 18 years to reach a yield of 200 MPa, or 13 years if the yield is 150 MPa. Distended clads younger than 13 years are observed, indicating that the tensile strength is lower than the compressive strength, as anticipated. These calculations include helium in both the porosity and the ullage outside the pellet.

<u>Calculation 2a</u>   determine fuel tensile yield as a function of the age at distension

If fuel age dictates moles of helium, and moles of helium and volume give a pressure, then the yield strength of the fuel in tension might be estimated from the time at which distension occurs. The yield strength in tension may be lower than the reported yield in compression. [13]

The range of ages among the known distended clads give a wide range of possible internal pressures. Some of the distended clads were discovered some time after distension occurred.

<u>Table 3.</u>

Distended clads		Age at discovery (years)	Age at venting (years)	LANMAS creation date	Clad mass measurement date	Encapsulation date
old	FC0009		19.4	9 Nov 1993		
old	FC0086		18.2	8 Dec 1994		
old	FC0201		17.3	11Jun 1997		
old	FC0312		16.8	11 Jun 1997		
hot	FC0371		10.1	08 Nov 2010	7/10/2003	3/20/2003
old	FC0421	5.7	5.7	30 Sept 2013	2/22/2008	8/1/2010
hot	FC0435	2.7	2.7	26 Aug 2010	9/28/2010	
hot	FC0437	2.7	2.7	26 Aug 2010	9/28/2010	
hot	FC0462		6.0			2/15/2013
hot	FC0463		6.0			2/15/2013

FC0421 is the best approximation to a threshold for the minimum age at which distension can be observed, without heating the clad. FC0421 was 5.7 years old at discovery, and had time to accrue 346 atm, suggesting that 346 atm is sufficient to distend the clad, in a volume consisting of pellet porosity and ullage. This gas loading would correspond to 35 MPa, suggesting a tensile strength of 35MPa. The range of ceramic tensile strengths is wide, ranging from alumina at 260 MPa and SiC at 310 MPa to graphite, at 4.8 MPa. Typically, the tensile strength of these ceramics is about 1/5 to 1/10 of the compressive strength [15] suggesting a value for the oxide tensile strength between 15 and 20 MPa (1/10) or 30 and 40 MPa (1/5). The value of 35 MPa is a reasonable tensile strength for the material.

In general, it is not known exactly when the distension occurred, and the age can be estimated only from the age at discovery, which hinders any effort to establish a timescale for distortion. In general, units that have expanded simply through the passage of time are older, between 16.8 and 19.4 years of age. These clads do not support the suggestion that the tensile strength of the fuel governs the onset of distension.

Clads that distended under high temperature excursions are excluded from the estimation of fuel tensile strength because the fuel does not fill the available space, and because the fuel may have fractured because of thermal stress, rather than because of gas pressure.

Tensile strength of the fuel is likely never reached, because insufficient pressure can accumulate. The vent covers on all distended clads measured to date have apparently opened, and the observed internal pressure in the clad is negligible. These observations indicate that it is unlikely that the tensile strength of the fuel is a factor in distension.

Calculation 3: distension governed by the estimated yield of the vent cover

The yield strength of the vent cover may govern distension, by allowing pressure to build up in the ullage and in the fuel until the vent cover yields, releasing the pressure. Once pressure in the ullage is released, either the helium inside the clad is vented, or the helium in the fuel may expand, increasing distension of the fuel.

All of the distended clads have a bulged vent cover, (Figure1) indicating that helium pressure has accumulated between the vent frit and the vent cover. The most likely behavior of the clads is gradual pressurization until the yield strength of the vent cover or vent cover weld is reached, and then self-venting of the clad. Every distended clad that has been examined has been shown to have negligible helium pressure inside, and also to contain air, indicating an open vent cover.

The dynamic yield of vent cover welds has been reported to occur at pressures between 150 psig and 180 psig. [2] These weld strengths were measured under dynamic pressurization, and probably provide a lower bound for the strength of the weld under gradual pressurization. Reaching a pressure of 150 psig in the available volume of 0.0048 liters, the porosity plus theoretical ullage, requires 6.84E-4 moles of gas at 600°C, which will accumulate in about 2 months. At 841°C, reaching this lowest yield pressure requires 5.36E-4 moles, or about 1.8 months' accumulation. To reach the higher yield pressure at 600°C requires more gas. To reach 180 psig requires 8.21E-4 moles and 2.5 months. At 841°C, reaching 180 psig requires 6.43E-4 moles which takes 2 months to accumulate.

These intervals around 2 months are shorter than the observed timescales for distension, even if the strength of the vent cover is higher under gradual pressurization than the measured values. Vent cover welds are expected to be only slightly stronger under static pressurization than in the dynamic pressurization measurements. [16]

Helium can be dissolved in the fuel. [6, 7, 8, 9] If the calculation of vent cover release pressure is repeated with 75% of the helium entrained in the fuel, then the vent cover weld rupture occurs at longer times. To accumulate 6.84E-4 moles and reach 150 psig requires 2.74E-3 moles if only 25% of the helium is in the gas phase. To get 2.74E-3 moles of gas at 600°C takes about 8 months, too short an interval to be consistent with observation of distension at 2.7 years. To reach 180 psig at 600°C with 8.21E-4 moles of gas exerting pressure would require 3.28 E-3 moles, which would accumulate in just over 9 months. This mechanism is only applicable if the clad remains at ambient temperature, allowing the gas to stay in solution in the fuel.

Once the vent cover weld is breached, the decrease in pressure inside the clad might precipitate release of gas from the fuel. When the pressure in the ullage drops, it is possible that helium entrained in the fuel is released inside the fuel, or that the helium exerts pressure inside the fuel where there was no pressure before, just as in thermal excursion. If gas accumulates in the solid, a mechanism for distension is possible without a thermal excursion. In reality the slightly porous vent cover may allow a gradual release, so that no abrupt cracking is observed in clads distended without a thermal excursion. A myriad of small cracks and a small fragment size in the aged clads are consistent with a slow rate of helium release. The helium release process is known to be sporadic and irregular, with instantaneous bursts and long releases over 2-3 minutes. [8] The helium is known to reside in a variety of sites within the oxide matrix, each releasing gaseous helium at a different energy. [6, 17]

Although vent cover weld rupture may play a part in distension of clads that get hot early in their lifetimes, distension is most likely not governed by the release of the vent cover, unless the timescale for detection is much longer than the timescale for actual distension. The opening and release of pressure by the vent cover does certainly occur, as indicated by gas sampling of these thermally perturbed clads. Determination of the actual vent cover release behavior as clads age would be a useful addition to the known aging behavior of these clads.

<u>Experimental measurements of vent cover behavior are feasible</u>

Determination of the actual vent cover release behavior as undistended clads age would be a useful addition to the known aging behavior of these clads. If the vent cover weld yields spontaneously after 2 or even 9 months, then there is no need to remove the vent cover after 18 months.

The vent cover serves to moderate the neutron dose produced by each GPHS, since it prevents admission of atmospheric oxygen to the clad that may allow back-exchange of oxygen isotopes with atmospheric oxygen. Even the small population of ^{17}O and ^{18}O in atmospheric oxygen will displace the carefully controlled ^{16}O composing the PuO_2 and allowing a rise in neutron output from the clad. [18]

Measuring the time between encapsulation and breach of the vent cover could be done without damage to the clads, by storing each clad in a closed vessel that could be sampled periodically using an RGA or a helium leak detector. If the back-exchange rate of the oxygen in the fuel was known, then periodic neutron emission rate (NER) measurements on the clads would serve to indicate the extent of exposure to atmospheric oxygen. Even without knowledge of the exact back-exchange rate, a rise in NER would strongly suggest an open vent cover. [18]

Experimental measurement of gas pressure in GPHS clads

The effectiveness of high temperatures in creating distension suggests an abrupt increase in helium output during thermal excursion. If helium pressure increases abruptly from the “entrained “ pressure to the “not entrained” pressure inside the clad, the clad may distend. Where there is insufficient gas pressure in the body of the clad for the vent cover weld to rupture, the fuel and the clad can accumulate substantial gas inventory before the vent cover opens spontaneously.

Experiments to measure the actual pressure inside GPHS clads as they age would provide a useful understanding of the entrainment of helium, the behavior of the vent cover, and the possible causes of distension. Measurement could provide a firm justification for venting GPHS at a particular age. If a new GPHS clad was vented, put in a small volume, and the pressure monitored at intervals, the helium release behavior of the fuel could easily be determined. The volume should be as small as possible to avoid imposing pressure drops on the fuel that might liberate helium that would otherwise dissolve in that fuel.

Summary

Several mechanisms by which helium could cause distension of the iridium cladding on a GPHS have been examined calculationally. Only a fraction of the expected inventory of helium is observed in the distended clads that have been gas sampled. Calculations indicate that all of the available volume in the clad is full of helium after a few days, or after 20 days if helium dissolves in the matrix. After this time, pressure may begin to build up inside the clad and inside the pellet, causing it to expand.

A calculation of the expansion as a function of the strength of the $^{238}PuO_2$ indicates that reaching the compressive strength of the solid requires a substantial inventory of gas, 12 to 45 years’ accumulation. While the compressive strength of $^{238}PuO_2$ is known, tensile strength is not. Assuming that tensile strength governs distension of the youngest unheated distended clad, 5.7 years old, a tensile strength of 35 MPa can be calculated, a reasonable value. However, observation of distended clads indicates that the vent over opens before pressures on these scales can accumulate.

Calculation of the pressure required to breach the vent cover weld suggests that the vent cover may give way after only 2 months, or may last as long as 9 months if helium dissolves in the PuO_2 matrix.

Experimental measurements of vent cover integrity and of the rate of helium evolution by the GPHS pellet would be useful in evaluating the potential for distension of GPHS clad, and in determining safe handling and storage practices and precautions. Helium gas evolution by the pellet may be less than anticipated, because of the potential for dissolution of helium in the PuO_2 matrix. [6, 7, 17]

Conclusions

Critical data bearing on the phenomenon that might be causing distension of the GPHS clads are lacking. Because vent covers are seldom removed on a clad that is retained and observed. Radiography is seldom performed on a clad except at the beginning of its life. The longevity of the vent cover weld is not known, nor is the pressure of free helium inside a GPHS clad as it ages. The tensile strength of the $^{238}PuO_2$ is not known. The actual time to distension is not known exactly, because observation of the GPHS clads in storage is sporadic.

Calculations suggest that none of the reasonable mechanisms proposed to explain distension occurs on the timescales suggested by the ages of the known distended clads, although the quantities supporting these calculations are often estimates. Neither the strength of the pellet, the strength of the vent cover weld, or the accumulation of helium pressure inside the clad appears to be responsible for distension of GPHS clads.

REFERENCES

1. J. Teague, "Integrity Evaluation of Swelled GPHS Clads" Los Alamos National Laboratory (15 October 2013)
2. J.P. Moore, "Quarterly Technical Progress Report of Radioisotope Thermoelectric Generator Materials Production and Technology Tasks for July through September 1995" Oak Ridge National Laboratory repot ORNUCF-95/88 (October 20, 1995)
3. Roberta N. Mulford, Diane Spengler, Jonathan Teague, Jared Mason, Paul D. Richardson, Jack Gower, and Joey Sanchez, "Report on Distended GPHS Clads, Initial Measurements and Mechanism" Los Alamos National Laboratory Report LA-CP-16-20293 (May 2016)
4. Tim George, private communication 2016
5. B.A. Mueller, D.D. Rohr, and R.N.R. Mulford, Los Alamos Report LA-5524, Los Alamos National Laboratory (1974.)
6. D. E. Peterson, J. W. Early, J. S. Starzynski, and C. C. Land, "Helium Release from Radioisotopic Heat Sources," Los Alamos National Laboratory Report LA-10023 (1984).
7. P. Angelini, R. E. McHenry, J. L. Scott, W. S. Ernst, Jr., and J. W. Prados, "Helium Release from $^{238}PuO_2$ Microspheres," Oak Ridge National Laboratory report ORNL-4507 (1970).
8. R.N.R. Mulford and B.A. Mueller, "Measurements of Helium Release from Materials Containing $^{238}PuO_2$," Los Alamos Scientific Laboratory Report LA-5215, (1973).

9. Helium Transport in PuO_2 as a Function of Temperature and Heating Rate. Roberta Mulford, Proceedings of the INMM 55th Annual Meeting, Los Alamos National Laboratory Report LA-UR-25325 (July 2014)
10. J. Teague, calculation of GPHS ullage, private communication (August 2019)
11. TA-55 Documented Safety Analysis TA55-DSA-2017-R0 (2017)
12. R.N. Mulford and D. E. Clark, "Distended GPHS Clads: Characterization of Fuel Particle Size," Los Alamos National Laboratory Report LA-UR-xxxx (26 August 2019)
13. M.G. Stout, R.W, Ellis, and R.A. Pereyra, "Mechanical Behavior of $^{238}UO_2$, $^{238}PuO_2$, and $^{239}PuO_2$ as a Function of Strain Rate and Temperature," Los Alamos National Laboratory Report LA-12811-MS (August 1994)
14. S. Wiederhorn, "Brittle Fracture and Toughening Mechanism in Ceramics," Annual Review of Materials Research 14(1):373-403 (November 2003)
15. https://www.ceramicindustry.com/ceramic-materials-properties-charts, accessed 15 September 2019.
16. Bo Song, Kevin Nelson, Helena Jin, Ronald Lipinski, John Bignell, G. B. Ulrich, and E. P. George, "Dynamic High-Temperature Characterization of an Iridium Alloy in Tension," Sandia Report SAND2015-8380 (September 2015)
17. C. Ronchi and J.P. Hiernaut, "Helium diffusion in uranium and plutonium oxides," J. Nuc. Mats. *325* pp. 1–12 (2004)
18. C. B. Chadwell and T. C. Elswick, "Neutron Emission Rate Reduction in PuO_2 by Oxygen Exchange," AEC Research and Development Report MLM-1844 (Sept 1971)